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14. ABSTRACT
This final report summarizes the progress achieved toward the primary goal of this project, single electron spin resonance measurement capability with atomic scale spatial resolution. While the primary goal of detection of the spin resonance of a single paramagnetic defect at room temperature has not been achieved yet, many advancements towards this goal have been made, as summarized in detail in previous annual progress reports. Here, a brief summary is made of these achievements. Additionally, a summary of the overall research findings to date and a description of the proposed next steps to achieve the primary goal are

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paramagnetic defects, spin detection, electron spin resonance, atomic scale spatial resolution, single electron tunneling

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Report Title

Final Report - Room temperature single-spin tunneling force microscopy for characterization of paramagnetic defects in electronic materials

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Enter List of papers submitted or published that acknowledge ARO support from the start of the project to the date of this printing. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

Received Paper

TOTAL:

Number of Papers published in peer-reviewed journals:

(b) Papers published in non-peer-reviewed journals (N/A for none)

Received Paper

TOTAL:

Number of Papers published in non peer-reviewed journals:

(c) Presentations

A. Payne, K. Ambal, C. Boehme and C.C. Williams, “Towards performing electron spin resonance measurements with a single spin at room temperature,” APS March Meeting, Baltimore, Maryland, March 18, 2013.

K. Ambal, A. Payne, D.P. Waters, C. Williams and C. Boehme, “Synthesis and Physical Characterization of think silicon dioxide layers with very high densities of E’ centers,” APS March Meeting, Baltimore, Maryland, March 18, 2013.

D.W. Winslow and C.C. Williams, “Creation of Electron Trap States in Silicon Dioxide By Local Electron Injection,” APS March Meeting, Boston, Massachusetts, February 28, 2012.

Number of Presentations: 3.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received	Paper

TOTAL:

Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received	Paper

TOTAL:

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):

(d) Manuscripts

Received		Paper
10/01/2013	1.00	K. Ambal, A. Payne , D. P. Waters, C.C. Williams, C. Boehme. Synthesis of thin silicon dioxide layers with high E' center densities and investigationof the E' center spin relaxation dynamics for single spin readout applications, Phys. Rev. B (submitted) (09 2013)

TOTAL: **1**

Number of Manuscripts:

Books

Received   Paper

TOTAL:

Patents Submitted

Single spin tunneling force microscope, provisional patent filed 6/15/2012.

Patents Awarded

Awards

Graduate Students

NAME	PERCENT_SUPPORTED	Discipline
Adam Payne	1.00	
FTE Equivalent:	**1.00**	
Total Number:	**1**	

Names of Post Doctorates

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Names of Faculty Supported

NAME	PERCENT_SUPPORTED	National Academy Member
Clayton Williams	0.08	No
Christoph Boehme	0.04	
FTE Equivalent:	**0.12**	
Total Number:	**2**	

Names of Under Graduate students supported

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: 0.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:...... 0.00

The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:...... 0.00

Number of graduating undergraduates who achieved a 3.5 GPA to 4.0 (4.0 max scale):...... 0.00

Number of graduating undergraduates funded by a DoD funded Center of Excellence grant for Education, Research and Engineering:...... 0.00

The number of undergraduates funded by your agreement who graduated during this period and intend to work for the Department of Defense 0.00

The number of undergraduates funded by your agreement who graduated during this period and will receive scholarships or fellowships for further studies in science, mathematics, engineering or technology fields:...... 0.00

Names of Personnel receiving masters degrees

NAME

Total Number:

Names of personnel receiving PHDs

NAME

Total Number:

Names of other research staff

NAME	PERCENT_SUPPORTED

FTE Equivalent:
Total Number:

Sub Contractors (DD882)

Inventions (DD882)

Scientific Progress

See attachment below

Technology Transfer

Final Report

Room temperature single-spin tunneling force microscopy for characterization of paramagnetic defects in electronic materials

Start date: 7/15/2010

PI: Clayton Williams, Department of Physics and Astronomy, University of Utah

Co-PI: Christoph Boehme, Department of Physics and Astronomy, University of Utah

April 8, 2014

Abstract

This final report summarizes the progress achieved toward the primary goal of this project, single electron spin resonance measurement capability with atomic scale spatial resolution. While the primary goal of detection of the spin resonance of a single paramagnetic defect at room temperature has not been achieved yet, many advancements towards this goal have been made, as summarized in detail in previous annual progress reports. Here, a brief summary is made of these achievements. Additionally, a summary of the overall research findings to date and a description of the proposed next steps to achieve the primary goal are given.

1. Table of Contents

2. Brief summary of research progress during the 3 year project

The goal of this research has been to develop a method to perform room temperature single-electron spin resonance measurements with atomic scale spatial resolution, using tunneling force microscopy. To achieve this goal, an electron must spontaneously shuttle back and forth (randomly telegraph) between a paramagnetic state at the surface of the sample and a paramagnetic state in the scanning probe microscope (SPM) tip. Under proper conditions, the force noise produced on the AFM cantilever changes when spin resonance conditions of one of the two states is reached. This measurable change in force detected noise on and off spin resonance provides the means by which atomic scale spin resonance measurements can be achieved.

Summarized below is a list of research achievements performed during the last 3 years. The achievements are explained in detail in each of the 3 previously submitted annual reports.

Year 1

1. Improvement of the force sensitivity of the UHV AFM system by approximately 1 order of magnitude. An external light source and fiber optic delivery system was constructed and integrated into the UHV AFM system. The AFM force detection system was then characterized, showing the signal to noise ratio was improved by approximately a factor of 10.

2. Quantitative simulations performed of the proposed single spin tunneling force measurement method. A detailed stochastic simulation of the random tunneling and spin flipping processes involved in the proposed single spin detection method was developed and performed. The simulation provides a detailed understanding of how the random telegraph noise changes when spin resonance conditions are reached. A quantitative electrostatic model was used to connect this ideal simulation to the measured force detected noise in the actual experiment. This allowed optimizing the measurement parameters for best signal to noise ratio [Pay2013,Pay2014].

3. Creation of trap states in silicon dioxide films by local AFM probe. The conditions under which trap states could be generated by applying short voltage pulses to an AFM probe tip positioned over a silicon dioxide film were studied. Evidence for the creation of trap states under the probe tip was obtained. [Win2012]

<u>Year 2</u>

4. Magnetic resonance system components were designed, constructed and integrated into the UHV AFM system. A custom RF coil and support was designed, constructed, calibrated and integrated into the UHV AFM system. The static magnetic field in the system was also calibrated for performing the single-spin magnetic resonance measurements.

5. Control and acquisition hardware and software was acquired and developed to perform single spin ESR measurements. A LABVIEW card was purchased and programed to control the sweep of the RF field frequency and to acquire as well as process the force noise detected during a magnetic resonance measurement.

6. First attempt at single spin ESR measurement was performed. A first attempt at performing the single-spin detection was made. System background noise variations were observed and corrections made to the system. No spin resonance signal was observed.

7. AFM probe tip induced trap states imaged by Dynamic Tunneling Force Microscopy line scans. Previous evidence for trap states created by AFM probe tip voltage pulses involved only single point measurements. Line scans of a single state produced under the AFM probe tip by a voltage pulse were obtained, showing the atomic scale localization of the trap states that were produced. [Win2012]

<u>Year 3</u>

8. System background and noise signal were characterized as the RF magnetic field frequency was swept. A study of the detection system background was performed. Several system components were improved, including the RF coil feed through and optical feed through, both of which cause back reflections. The RF feed through produces frequency-dependent modulations in the RF power at the coil and the optical feed through can cause laser power instabilities.

9. Observation of the random telegraph noise in force measurements between tip oxide and sample oxide as a function of tip-sample gap and applied voltage. Many tunneling measurements were performed with oxidized AFM probe tips near oxide sample surfaces. These measurements were performed as a function of applied voltage and gap. The results showed that the observed voltage and gap dependence was not consistent with an electron trap state to electron trap state tunneling model. This raised questions about the origin of the random telegraph signal observed in the AFM measurements. Combined with the observation that the telegraph noise occurs with a high probability, it was concluded that the observed telegraph noise was likely associated with an atomic bonding instability that occurs when the two oxide surfaces (of same composition) come close enough that spontaneous chemical bonding may occur. **The observations indicate that force variations may occur which are associated with random fluctuations in dangling bonds or atoms at the surface of the two oxides.** In some cases, the bonding events could be bistable, producing a random telegraph signal. This new picture does not preclude the fact that random telegraph signal like force variations could be produced by a single electron tunneling between two trap states, but it may mask the presence of such events.

10. Quantitative AFM noise measurements have been performed with oxidized AFM probe tips and oxide surfaces. These measurements show that the sensitivity of the proposed AFM approach is adequate to perform single electron ESR measurements at room temperature under typical conditions, if the atomic bonding instability can be eliminated [Pay14].

3. Summary of overall research

During this three year project, a number of advancements have been made toward single spin detection, however, the goal of demonstrating room temperature single spin detection has not been achieved. Improvements to the UHV AFM system force sensitivity have been made and noise measurements have shown the sensitivity of the system to be adequate to achieve the single spin ESR goal. An additional source of signal/noise has been observed - a random telegraph signal that could be caused by binary bonding events between the tip and sample oxide surfaces. It is believed that these events are masking the electron tunneling signal need to achieve the primary goal.

4. Proposed research toward single spin ESR detection

There are several possible trajectories toward achieving single spin ESR detection.

1. Find ways to eliminate the instability that exists between the tip and sample oxide surfaces. This is challenging because the probe tip oxide surface would have to be chemically modified to produce a surface that would not strongly interact with the sample oxide. Alternatively, one could replace the tip oxide with another material that has little or no affinity for silicon dioxide, but there are few materials that would be expected to have paramagnetic defects with long spin-relaxation times at room temperature. This is not the preferred direction for future research.

2. Move the experiment to low temperature and use phosphorus donors in silicon as the paramagnetic probe tip state (with the usual paramagnetic dangling bond states (E' or P_b centers) in the oxide sample. This solves several problems. Firstly, silicon AFM probe tips have already been used many times to stably image oxide surfaces without the bonding instability.

Secondly, silicon probe tips can be doped or implanted with phosphorus donor atoms at a high enough density that the likelihood of finding a phosphorus donor atom within a fraction of a nanometer of the surface is high. Thirdly, phosphorus is a well understood paramagnetic state which has a very long spin-relaxation time at low temperature [Tyr2003, Tyr2012]. Forth, our research group has already studied a silicon – silicon dioxide system by conducting-AFM (c-AFM). In this study, clear evidence that pairs of single dangling bond states (believed to be P_b centers at the silicon/silicon dioxide interface) and single phosphorus atoms can be addressed by the c-AFM. Under these conditions, the current through these two paramagnetic pairs is spin dependent. The current through the pair is larger when the spins are oppositely aligned. Under magnetic resonance conditions (for one of the two states), both, the average current through the pair and the noise on that current will differ than off-resonance. This provides a means to directly detect the spin resonance or g-factor of a single paramagnetic state.

Once such a measurement is performed, the same system can then be used to incrementally approach the measurements originally proposed and worked on for the last three years. To do this, electrically isolated phosphorus donor atoms at the AFM probe tip apex (at low temperature) will be brought within tunneling range of electrically isolated P_b centers. Voltages will be applied to bring the energy of the singly occupied phosphorus state into alignment with the energy of the doubly occupied P_b state. Under this condition, the electron from the phosphorus state will randomly tunnel back and forth between the two states, showing the exact behavior that was originally proposed (using two E' center states). The same analysis developed in the last three years can then be applied to this new system. The expected result will be single spin ESR detection with atomic special resolution. Once this demonstration has been achieved and single spin detection at low temperatures is available, substitutes for the phosphorous probe spins could be explored that work at higher temperatures.

5. Final Project Report Summary

This final report summarizes the progress achieved toward room temperature single electron spin resonance measurements with atomic scale spatial resolution. The advancements made during the project have been briefly described and a summary provided which describes what has been learned during the project. A future research direction to achieve the primary goal of single spin ESR detection with atomic spatial resolution has been proposed.

6. References

[Pay2013] A. Payne, K. Ambal, C. Boehme and C.C. Williams, “Towards performing electron spin resonance measurements with a single spin at room temperature,” APS March Meeting, Baltimore, Maryland, March 18, 2013.

[Pay2014] A. Payne, K. Ambal, C. Boehme and C.C. Williams, “A concept for room temperature single-spin tunneling force microscopy with atomic resolution,” to be submitted (May, 2014) to Nature Nanotechnology.

[Tyr2003] Alexei M. Tyryshkin et al., Electron spin relaxation times of phosphorus donors in silicon, Phys. Rev. B 68, 193207 (2003).

[Tyr2012] Alexei M. Tyryshkin et al., Electron spin coherence exceeding seconds in high-purity silicon, Nature Materials, 11, 143–147 (2012).

[Win2012] D.W. Winslow and C.C. Williams, “Creation of Electron Trap States in Silicon Dioxide By Local Electron Injection,” APS March Meeting, Boston, Massachusetts, February 28, 2012.